High Concentration NOx Removal Using a Multistage Combustor

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The U.S. Department of Energy's (DOE) Transuranic and Mixed Waste Focus Area has contracted MSE Technology Applications, Inc. (MSE) to perform this test project at the DOE's Western Environmental Technology Office in Butte, Montana.

For many years, the Idaho National Engineering and Environmental Laboratory (INEEL) has used a calciner process, beginning with the Waste Calciner Facility (WCF), to treat sodium-bearing radioactive and hazardous liquids to convert millions of gallons of liquids into a more stable solid form. The waste is located at the Idaho Nuclear Technology and Engineering Center (INTEC), previously referred to as the "Chemical Plant." The WCF was replaced in 1982 with the New Waste Calciner Facility (NWCF). Since 1982, many hazardous waste and air quality laws have been enacted that affect the operation of the NWCF. The most recently enacted regulatory driver is the U.S. Environmental Protection Agency's Maximum Achievable Control Technology (MACT) rule, and addressing the requirements of the MACT rule is the primary reason for this test project.

As a result of thermal processing of high nitric acid (HNO_3)-laden waste, offgas from the NWCF contained high levels of oxides of nitrogen (NO_x), which were problematic to future necessary compliance sampling and caused an unsightly orange cloud within the site area. After evaluating available technologies, including selective catalytic reactors, a multistage combustion NO_x removal system was selected by the NWCF as the technology most feasible to control NO_x emissions. MSE awarded a contract to the John Zink Company, which provided a multistage combustion NO_x removal system (NO_xidizer™) for testing.

An Environmental Impact Statement (EIS) draft recently completed implies that the NWCF, now indefinitely inactive, will likely be decommissioned. A melter (joule heater) waste processing technology is one technology favored by the EIS as an alternative to the NWCF. Since June 2000, the focus of this test project shifted from using the NWCF calcifier process to direct vitrification by a melter to treat the target waste. Since the waste will still contain a high HNO_3 component, its melter waste treatment technology will likely also have high NO_x emissions and will require an NO_x abatement system for the same reasons that the NWCF required an NO_x abatement system. The current melter conceptual design calls for a NO_xidizer™ immediately

downstream of the melter (but also upstream of a scrubber), a mercury sorbent bed, and a variety of other equipment.

The NO_xidizer™ is a three-stage combustor. The first stage converts waste gas NO_x under reducing conditions at 2,100 ^{o}F to nitrogen and oxygen. The second-stage water quenches the waste gas to 1,550 ^{o}F, and the third stage oxidizes the remaining combustibles (carbon monoxide, hydrogen, and miscellaneous hydrocarbons). During the spring of Fiscal Year 2000, the natural gas-fired NO_xidizer™ was tested by MSE. Results show the technology is a viable process for treating high levels of NO_x, up to 40,000 parts per million volume (ppmv) in offgas.

The primary objective of this test was to investigate the NO_xidizer's NO_x destruction removal efficiency (DRE), this time using propane as a fuel, while removing NO_x from a wastestream carrying up to 40,000 ppmv NO_x. Previous natural gas-fueled testing serves as a baseline. The unit was converted to propane because that is the fuel expected to be used at INTEC. Additional objectives while operating the optimized NO_xidizer™ NO_x DRE settings were to determine the total hydrocarbon (THC) baseline emissions of the NO_xidizer™, determine the NO_xidizer's THC DRE while injecting an organic liquid into the wastestream, measure the NO_xidizer's speciated NO_x levels downstream of the first-stage reducing section and in the stack, and inject a mix of elemental and speciated mercury into the NO_xidizer's wastestream and sample the NO_xidizer's stack to determine the outcome of the mercury.

Secondary objectives were to perform oxygen-enrichment tests and inject a dioxin/furan surrogate into the NO_xidizer's wastestream while drawing manual samples to determine the NO_xidizer's DRE for dioxins/furans, funding permitting. Due to budget limitations the secondary objectives were not met.

Most of the test objectives for the propane-fueled NO_xidizer™ test project were met. At the optimized conditions, NO_x DRE was up to 6% and approached 97% of the theoretically possible DRE, with an inlet NO_x level of 50,000 ppmv, wet basis. Benzene was used as a THC surrogate and was spiked at a concentration of 370 ppmv in the NO_xidizer™ waste gas. No detectable benzene was found at the NO_xidizer™ stack, which implies the NO_xidizer™ system achieved at least 99.7% organic destruction.

Limited data (approximately 10 minutes of continuous emissions monitoring system data) was obtained for NO_x species just downstream of the reducing section and at the stack. This data showed there was an average of 35 ppmv of nitrogen oxide, 70 ppmv of HNO_3, HNO_3 slip, and no detectable nitrogen dioxide, all expressed on a wet basis. Comparing this data with measurements at the stack outlet indicate that NO_x reformation downstream of the reducing section is small—approximately 400 ppmv wet basis. Due to limited data, this statement should be considered as very qualitative indicator only.

A mix of speciated mercuric chloride ($HgCl_2$) and elemental mercury was spiked into the waste gas feed at a combined concentration of 4,100 micrograms per dry standard cubic meter (µg/dscm). Approximately 70% of the mix was $HgCl_2$, and 30% of the mix was elemental mercury. Mercury samples taken at the NO_xidizer™ stack indicated that over 99% of the mercury present is elemental. This result verified the volatilization temperatures 570 ^{o}F for

$HgCl_2$ and 675 ^oF for elemental mercury and the thermal breakdown temperature, 1500 ^oF , for $HgCl_2$. No appreciable effects on NO_x DRE were observed during the mercury injection testing.

Follow-on testing will involve examining ways to reduce total mass flow of the system, most likely by oxygen enrichment of combustion air in the reducing section.

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